Generation and amplification of Raman Stokes and anti-Stokes waves

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Compiled February 1, 2008

We present general analytical expressions of Stokes and anti-Stokes spectral photon-flux densities that are spontaneously generated by a single monochromatic pump wave propagating in a single-mode optical fiber. We validate our results by comparing them with experimental data. Limiting cases of the general expressions corresponding to interesting physical situations are discussed.

OCIS codes: 190.5650, 190.4380, 190.5890, 270.5530

The third order nonlinearity seen by light propagating in optical fibers is composed of two parts: a quasi instantaneous contribution coming from the electronic response, and a delayed Raman response coming from the coupling to molecular vibrations (phonons). The influence of these two terms on the propagation of light are often considered separately, see for instance the treatment in [1]. But in many cases they must be treated collectively, together with the influence of dispersion which plays an essential role, particularly when the phase matching conditions are (even approximately) satisfied. That a holistic approach is necessary was first understood by Bloembergen and Shen [2], and confirmed experimentally in later work. For instance it was shown that the gain of the Raman Stokes wave is strongly affected by the dispersion, and can even vanish when there is zero phase mismatch [3, 4]. In addition it was shown in [2], and confirmed experimentally in [5], that the Raman anti-Stokes wave, which in the naive approach is exponentially damped, in fact grows exponentially at late times. This effect has important implications for supercontinuum generation in photonic crystal fibers using long pump pulses (ps and longer). In these systems spontaneous amplification of vacuum fluctuations at the anti-Stokes wavelengths can give rise to a blue detuned supercontinuum [6], see [7, 8] for further studies and [9] for a review of supercontinuum generation.

A quantum description of light propagation in optical fibers, including both the instantaneous Kerr non linearity and the Raman scattering has been developed [10] and used to quantify noise limits on squeezing [11], noise in $\chi^{(3)}$ parametric amplifiers [12], noise in coherent anti-Stokes Raman scattering [13], and noise in photon pair generation experiments [14, 15]. In the present work we apply this quantum theory, using the formulation of [12], to describe the spontaneous growth of Raman Stokes and anti-Stokes waves from vacuum fluctuations. Our analytical predictions are compared to experimental results reported in [5, 6] which had previously been fitted with semi-empirical formulae, or numerical calculations.

We consider a continuous pump with complex amplitude $\sqrt{P}\ \mathrm{e}^{i\phi(x,t)}$, where P is the power flowing through the fiber and $\phi(x,t) = -\omega_p t + [k_p + \gamma P]x$. In this expression, ω_p is the pump frequency and k_p is the (linear) wave number. Self-phase modulation gives a nonlinear contribution γP to the total wave number. Here γ is the third-order nonlinearity parameter of the fiber [1]. The quantum field operator

$$A(x,t) = \mathrm{e}^{i\phi(x,t)} \int_0^\infty \left(\sqrt{\frac{\hbar\omega_s}{2\pi}}\ A_s(\Omega,x)\ \mathrm{e}^{i\Omega t} + \sqrt{\frac{\hbar\omega_a}{2\pi}}\ A_a(\Omega,x)\ \mathrm{e}^{-i\Omega t} \right) \mathrm{d}\Omega + \mathrm{h.c.} \tag{1}$$

describes the perturbations around this stationary solution through the combined effect of Raman scattering and four-wave mixing. These are composed of symmetrically detuned Stokes and anti-Stokes photons with respective frequencies $\omega_s = \omega_p - \Omega$ and $\omega_a = \omega_p + \Omega$, and wave numbers k_s and k_a. The operators $A_s(\Omega,x)$ and $A_a(\Omega,x)$ are *destruction operators* for Stokes and anti-Stokes photons. Their equations of motion can be deduced from the quantum theory of light propagation and solved analytically (so long as the perturbation field is small compared to the pump) [12].

Once $A_s(\Omega,x)$ and $A_a(\Omega,x)$ are known, any physical quantity can be computed. Here we are concerned by the mean *spectral photon-flux densities*

$$f_{s,a}(\Omega,x) = \lim_{\epsilon\to 0} \frac{1}{2\pi\epsilon} \times \int_{\Omega-\frac{\epsilon}{2}}^{\Omega+\frac{\epsilon}{2}} \int_{\Omega-\frac{\epsilon}{2}}^{\Omega+\frac{\epsilon}{2}} \langle A^\dagger_{s,a}(\Omega_1,x) A_{s,a}(\Omega_2,x)\rangle \mathrm{d}\Omega_1 \mathrm{d}\Omega_2. \tag{2}$$

Using the approach of [12], we find

$$f_s(\Omega,x) = \frac{1}{2\pi}\ \frac{|\chi(\Omega)|^2}{|\kappa(\Omega)|^2}\ |\sinh(\kappa(\Omega)x)|^2 + \frac{|\mathrm{Im}[\chi(\Omega)]|}{\pi}\ \rho_+(\Omega,x)\ \ (n(\Omega)+1) \tag{3a}$$

$$f_a(\Omega,x) = \frac{1}{2\pi}\,\frac{|\chi(\Omega)|^2}{|\kappa(\Omega)|^2}\,|\sinh(\kappa(\Omega)x)|^2 + \frac{|\mathrm{Im}[\chi(\Omega)]|}{\pi}\,\rho_-(\Omega,x)\;n(\Omega), \tag{3b}$$

with

$$\rho_\pm(\Omega,x) = \int_0^x \mathrm{d}x' \left|\cosh(\kappa(\Omega)x') \pm i\frac{\Delta k(\Omega)}{2\kappa(\Omega)}\sinh(\kappa x')\right|^2 . \tag{4}$$

The photon fluxes depend on three basic parameters: the pump power P, the detuning Ω and the fiber temperature T. The pump power and the nonlinear response of the fiber are combined as $\chi(\Omega) = \gamma P\,[(1-f_r) + f_r\chi_r(\Omega)]$, where the nonlinearity is decomposed into an instantaneous (electronic) part and a retarded (molecular or Raman) one. $f_r \approx 0.18$ is the fraction of the total nonlinearity due to the Raman scattering and $\chi_r(\Omega)$ is the normalized spectral Raman response ($\mathrm{Re}[\chi_r(0)] = 1, \mathrm{Im}[\chi_r] < 0$). The complex parameter $\kappa(\Omega) = [-(\Delta k(\Omega)/2)^2 - \Delta k(\Omega)\chi(\Omega)]^{1/2}$ controls the growth rate of the Stokes and anti-Stokes waves. It depends both on $\chi(\Omega)$ and on the linear phase-mismatch $\Delta k(\Omega) = k_s + k_a - 2k_p$. When the fiber is pumped sufficiently far away from the zero dispersion wavelength, the phase-mismatch $\Delta k(\Omega)$ is well approximated by $\beta_2\Omega^2$, where β_2 is the group-velocity dispersion parameter. The real part of $\kappa(\Omega)$ is chosen positive so that it can be interpreted as the *amplification gain* for small signals at frequencies $\omega_p \pm \Omega$. Finally, the temperature T influences the photon fluxes through the factor

$$n(\Omega) = (\exp(\hbar\Omega/k_BT) - 1)^{-1} \tag{5}$$

related to the *phonon population*.

The integral in Eq. (4) can be carried out exactly, leading to somewhat cumbersome expressions. Here however, we prefer to focus on two physically important limits : the *spontaneous scattering limit* ($|\kappa|x \to 0$) and the *stimulated amplification and wave-mixing limit* ($\mathrm{Re}[\kappa]x \to \infty$). The first one is of great importance for photon-pair generation, while the second one applies to supercontinuum generation.

Considering the limit of small $|\kappa|x$, Eqs. (3) give

$$f_s \approx \frac{1}{\pi}|\mathrm{Im}[\chi]|\;x\;(n+1) \tag{6a}$$

$$f_a \approx \frac{1}{\pi}|\mathrm{Im}[\chi]|\;x\;n, \tag{6b}$$

up to the first order in $|\kappa|x$. This contribution to the total photon flux is called the *spontaneous Raman scattering.* Stokes and anti-Stokes photons are emitted independently. A Stokes photon emission is always accompanied by the emission of a phonon of energy $\hbar\Omega$, in contrast to the anti-Stokes process for which conservation of energy requires a phonon to be absorbed. For this reason, the anti-Stokes process in inhibited when the phonon population is zero (low temperature limit). Using Eq. (5), one finds that $\lim_{|\kappa|x\to 0} f_a/f_s = \exp[-\hbar\Omega/(k_BT)]$ in accordance with the usual formulation theory of spontaneous Raman scattering in fibers (see [5] and references within). Upon adding the extra term $|\chi|^2x^2/(2\pi)$ to Eqs. (6a) and (6b) one accounts for the *spontaneous four-photon scattering* process in which a Stokes photon and an anti-Stokes photon are emitted together after the absorbtion of two pump photons. This is the process used for photon-pair generation in fibers. Since it is only of second order in $|\kappa|x$, spontaneous Raman scattering always plays a detrimental role in photon-pair generation experiments and is referred to "Raman noise" (see [14, 15] for a more thorough discussion). It is interesting to note that linear dispersion has no impact on the values of the photon fluxes up to the second order in $|\kappa|x$ since they do not depend on Δk. However, the validity of the Maclaurin expansion is restricted to small values of Δk because $|\kappa| \to (\Delta k/2)$ for fixed pump power and large Δk values.

The asymptotic behavior for $\mathrm{Re}[\kappa]x \to \infty$ is also simple since we keep only the exponentially growing terms in Eqs. (3) :

$$f_s \sim \frac{\mathrm{e}^{2\mathrm{Re}[\kappa]x}}{8\pi}\left(\frac{|\chi|^2}{|\kappa|^2} + \frac{|\mathrm{Im}[\chi]|}{\mathrm{Re}[\kappa]}\frac{|\kappa + i\frac{\Delta k}{2}|^2}{|\kappa|^2}(n+1)\right) \tag{7a}$$

$$f_a \sim \frac{\mathrm{e}^{2\mathrm{Re}[\kappa]x}}{8\pi}\left(\frac{|\chi|^2}{|\kappa|^2} + \frac{|\mathrm{Im}[\chi]|}{\mathrm{Re}[\kappa]}\frac{|\kappa - i\frac{\Delta k}{2}|^2}{|\kappa|^2}\;n\right). \tag{7b}$$

Note that $|\kappa + i\frac{\Delta k}{2}| > |\kappa - i\frac{\Delta k}{2}|$ which implies that the Stokes wave is always stronger than the anti-Stokes wave, as expected. In Fig. 1 we have plotted the ratio f_a/f_s of the anti-Stokes to Stokes fluxes at the peak of the Raman gain ($\Omega/(2\pi) = 13.2$ THz) at $T = 300$ K as a function of $\gamma P/\Delta k$. In these circumstances, $n = 0.14$ and $\chi = \gamma P\;(0.82 - i\;0.25)$ for silica optical fibers. When $\gamma P/|\Delta k| \gg 1$ pair creation dominates over Raman scattering in Eqs. (7), both in the normal and anomalous

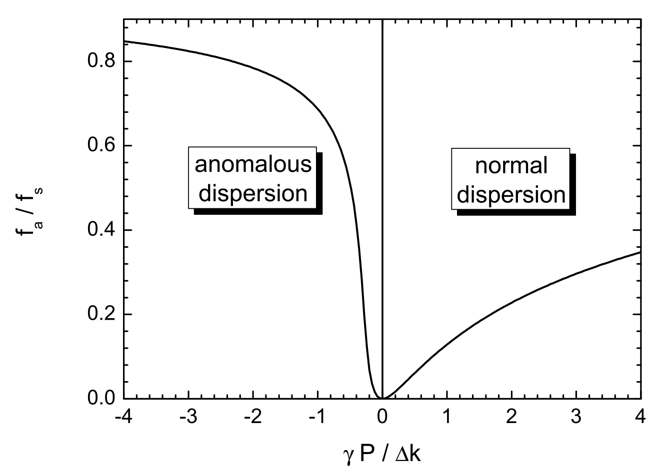

Fig. 1. Ratio of anti-Stokes to Stokes photon fluxes, at the peak of the Raman gain, as a function of $\gamma P/\Delta k$.

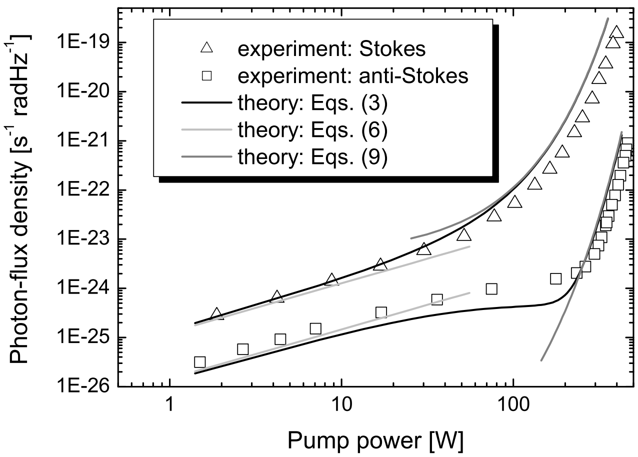

Fig. 2. Evolution of the Stokes and anti-Stokes spectral photon-flux densities with the pump power P. The parameters are: $\gamma = 23\ \mathrm{W}^{-1}\mathrm{km}^{-1}$, $x = 2.85$ m, $\beta_2 = 50\ \mathrm{ps}^2\mathrm{km}^{-1}$ and $T = 293$ K. (Experimental data from [5].)

dispersion regimes, and we have

$$f_s \simeq f_a \sim \frac{\mathrm{e}^{2\mathrm{Re}[\kappa]x}}{8\pi}\frac{|\chi|^2}{|\kappa|^2}. \tag{8}$$

However f_a/f_s tends much faster to 1 in the anomalous dispersion regime than in the normal dispersion regime, as is apparent from Fig. 1. When the opposite limit $\gamma P/|\Delta k| \ll 1$ is considered, the approximation $\kappa \simeq |\mathrm{Im}[\chi]| + i(\Delta k/2 + \mathrm{Re}[\chi]) + o(|\chi/\Delta k|)$ holds and

$$f_s \sim \frac{\mathrm{e}^{2|\mathrm{Im}[\chi]|x}}{2\pi}\ (n+1) \tag{9a}$$

$$f_a \sim \frac{\mathrm{e}^{2|\mathrm{Im}[\chi]|x}}{2\pi}\ \frac{|\chi|^2}{\Delta k^2}\ (n+1). \tag{9b}$$

The ratio of anti-Stokes to Stokes fluxes is then given by $f_a/f_s = |\chi|^2/\Delta k^2$. It is worth noting that it is *independent of temperature*. This simple ratio was first exhibited in [5]. However, it should be noted that its origin is more complicated than the argument given in [5] since the different processes of pair creation and phonon emission/absorption all contribute to it.

Even though Eqs. (7), (8) and (9) are valid for arbitrary detunings Ω, we now focus on $\Omega/(2\pi) = 13.2$ THz (corresponding to the Raman peak) in order to compare theory to experiments.

Experimental Stokes and anti-Stokes spectral photon-flux densities in the $\gamma P/|\Delta k| \ll 1$ regime are available from [5]. They are plotted in Fig. 2 as a function of the pump power P. For the pump powers that are considered $\gamma P/|\Delta k| \leq 0.032$. As seen from the figure, the asymptotic formula (9) [dark grey curves] apply for $P \geq 300$ W only. At lower pump power the condition $\mathrm{Re}[\kappa]x \gg 1$ is no longer satisfied. For very low pump powers, one can however apply the formula (6) [light grey curves] since $|\kappa|x \ll 1$. Outside these limiting cases Eqs. (3) reproduce very well the spectral photon-flux densities that are observed experimentally [black curves].

In [6], experimental data are obtained for a photonic crystal fiber with $\gamma = 150\ \mathrm{W}^{-1}\mathrm{km}^{-1}$ and $\beta_2 = 7\ \mathrm{ps}^2/\mathrm{km}$ (normal dispersion). Measurements have been carried out with $x = 3$ m and $P = 90$ W, as well as with $x = 0.7$ m and $P = 400$ W. In both cases $\mathrm{Re}[\kappa]x \gg 1$ but since $\gamma P/|\Delta k|$ is close to one neither Eqs. (8) nor Eqs. (9) can be used to compute the ratio f_a/f_s. Our result (7) should be used instead. It predicts the ratio $f_a/f_s = 0.028$ for $P = 90$ W and $f_a/f_s = 0.16$ for $P = 400$ W ($T = 300$ K). These are in good agreement with the measured ratios ($f_a/f_s = 0.016$ and 0.22 respectively) given the uncertainty on the experimental parameters in [6] and probable polarization effects.

In summary we have developed an analytic theory to account for the growth of the Stokes and anti-Stokes waves from the combined effects of pair creation and Raman scattering. The results are in good agreement with earlier experimental observations. They should find applications in the optimization of supercontinuum sources based on long pump pulses.

This research was supported by the EU project QAP (contract number 015848) and the IAP programme, Belgium Science Policy, under grant P6-10. We thank S. Coen and D. A. Wardle for sharing with us their experimental data.

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